The corresponding linewidth is given by

$$\sigma^2(q) = q^2(\tfrac{1}{2}\delta + \tfrac{1}{2}\Gamma^{-1} + \tfrac{2}{45}\Gamma^{-1}U/NkT)\omega_p{}^2.$$

Both U/NkT and δ are known from the computations of equilibrium properties.[1] U/NkT is always negative; δ is negative at $\Gamma = 1$, but becomes positive beyond $\Gamma \simeq 3$. Thus the coefficient of q^2 in (4) turns out to be positive for $\Gamma \lesssim 3$, and negative at higher Γ, in agreement with our observations. The high-Γ dispersion curve is not unlike the "optical" (longitudinal) phonon branch in the crystalline OCP.[1] This suggests that at high Γ "shearing" modes, related to the transverse motions, become important. This point is presently under investigation.

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Direct Spectroscopic Observation of Electrons in Image-Potential States Outside Liquid Helium

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We present the first direct spectroscopic study of electrons bound in image-potential–induced surface states outside liquid helium. We have observed absorption lines which show the expected linear Stark effect in the presence of an electric field. Zero-field splittings extrapolated from the data are 125.9 and 148.6 GHz for electronic transitions from the ground state to the first two excited states in the image-potential well.

The potential energy of an electron outside liquid helium is composed of two parts: (1) a long-range classical image potential, and (2) a short-range repulsive barrier at the surface. The latter is basically due to the exclusion principle, which forces the wave function of an extra electron to be orthogonal to those of the two electrons already present on each helium atom. The possibility that such a potential could cause surface states outside liquid helium was first noted by Sommer.[1] The idea was independently developed and further elaborated a few years later by Cole and Cohen,[2] and by Shikin.[3] Initial experimental investigations designed to measure the mobility[4] and lifetime[5] were apparently refuted by Ostermeier and Schwarz.[6] However, a cyclotron-resonance study by Brown and Grimes[7] showed the existence of surface states but gave no information about the potential. We are now presenting a direct spectroscopic study which shows conclusively the presence of an image-potential–induced surface state which displays a first-order linear Stark effect. In addition we are able to study the behavior of the image potential at the liquid-helium surface.

Our experiment consists of observing the absorption of microwave radiation by electrons on the surface of liquid helium. This is accomplished by tuning the splitting between the eigenstates in the image-potential well with an electric field until it is in resonance with our applied frequency. Observations are made at various applied frequencies and the unperturbed resonant frequencies are deduced by extrapolation to zero electric field.

We shall now briefly review the simple theory

of these states before presenting the details of our experiment. Since the valence electrons in helium are so tightly bound, a classical electrostatic image potential is accurate even as close to the surface as a few angstroms. Thus we expect $V(x) = -Ze^2/x$, where $Z = (\epsilon_0 - \epsilon_g)/4(\epsilon_0 + \epsilon_g)$; ϵ_0 and ϵ_g are the dielectric constants for liquid and gaseous helium, respectively, and x is the distance from the surface. Putting in the known values of $\epsilon_0 = 1.0572$[8] and $\epsilon_g - 1 < 10^{-4}$ we obtain $Z = 6.95 \times 10^{-3}$ for all temperatures below 1.4 K.

Assuming the surface to be flat allows one to separate the wave function for perpendicular and parallel motion. This yields plane waves parallel to the surface and an equation

$$(\hbar^2/2m)d^2\Phi/dx^2 + (E + Ze^2/x)\Phi = 0 \quad (1)$$

for perpendicular motion outside the helium. Inside the helium we assume the potential to be a constant equal to the measured barrier height of 1 eV.[9] Equation (1) is identical to the equation for $rR_{n0}(r)$, where $R_{n0}(r)$ is the nth radial solution with $l = 0$ for the hydrogen atom. Thus, the characteristic energies of the perpendicular part of the surface states will be approximately $Z^2\mathcal{R} \sim 1$ meV (~240 GHz), where $\mathcal{R}$ is the Rydberg. Since these energies are much less than the barrier height, a good approximation is to assume that the wave function vanishes at the surface, leading to a "hydrogenic" energy spectrum $E_n = (-Z^2/n^2)\mathcal{R}$ which describes our data to 5%. In this approximation the first three eigenstates are 159.0, 39.8, and 17.7 GHz below the vacuum. The splittings between the ground state and the next two are thus 119.2 and 141.3 GHz. The characteristic length or "Bohr radius" for this system is $a_0/Z = 76$ Å. The values of $\langle x \rangle$ for the three lowest eigenstates are then 114, 456, and 1026 Å, respectively.

The effect of an electric field $\vec{\mathcal{E}}$ in this approximation can be easily computed by first-order perturbation theory. Since the perturbing potential is simply $e\mathcal{E}x$, each state is shifted in energy by $e\mathcal{E}\langle x \rangle$. Thus, we expect the linear Stark shifts between the ground state and the first and second excited states to be 0.8 and 2.1 GHz $(\mathrm{V/cm})^{-1}$, respectively.

Our experimental apparatus consists of a right circular cylindrical absorption cell of radius 1.1 cm and height 0.32 cm. Microwaves are coupled into and out of the cell by two tapered guides oppositely placed on the side walls. One leads to a standard waveguide which carries microwaves from the source (a Si point-contact diode doubler driven by an M-band klystron) to the cell. The other is terminated by our detector, an InSb bolometer[10] which is operated in the liquid helium. Electrons are supplied by briefly heating a 0.001-cm-diam thoriated tungsten filament. The top and bottom surfaces of the cell are electrically isolated from the sides which are grounded to the incoming waveguide. The cell is placed in a vacuum-tight can which is then immersed in a pumped helium bath capable of cooling to 1.2 K.

A typical experimental run consists of condensing sufficient helium into the vacuum can to fill half the cell at 1.2 K. The voltages on the top and bottom plates are adjusted so that the helium surface is at a small positive potential. The center of the filament is biased so that electrons are attracted only toward the helium surface and cannot reach the top plate. The filament is then briefly heated to charge the helium surface with electrons until their space-charge potential cuts off further current to the surface. The filament is then turned off and the voltage between the top and bottom plates is swept by a triangular wave form applied in such a way as to keep the helium surface at the same positive potential. This procedure enables us to vary the perpendicular electric field on the electrons without allowing the number on the surface to change. (Charge can be held on the surface for hours!) Simultaneously with the above potentials, a small 50-kHz sine-wave voltage is applied across the plates through a transformer. The bolometer output is synchronously demodulated at 50 kHz, yielding dA/dV, the derivative of the microwave absorption with respect to the voltage across the cell. The absorption derivative is fed into one axis of an x-y recorder while the dc potential difference between the plates is applied to the other axis. Some data were also taken using a Fabri-Tek signal averager.

Figure 1 shows an experimental trace taken at 160 GHz. The larger absorption-derivative signal is due to transitions from the ground state, $n = 1$, to the first excited state, $n = 2$; the smaller signal arises from transitions between the ground state and the second excited state, $n = 3$. (We have also observed a very weak line we attribute to a transition to the third excited state.) Both lines display a small systematic shift in position with increasing surface charge density except when the cell is exactly half full.[11] By taking several traces at different surface charge densities in the range 10^6 to 10^7 electrons/cm^2 and extrap-

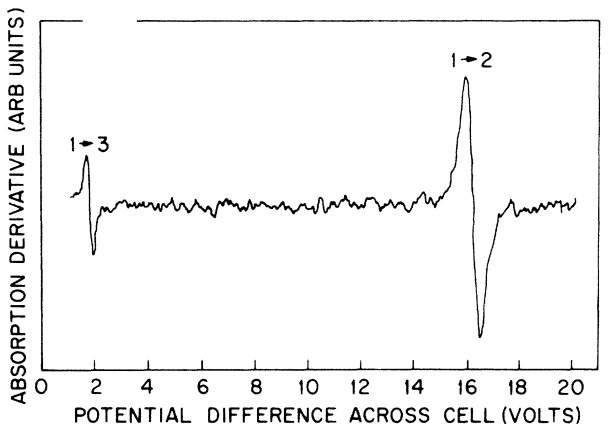

FIG. 1. Experimental trace showing absorption derivative versus voltage across the experimental cell at a frequency of 160 GHz and temperature of 1.2 K. The linear Stark effect is utilized to tune the splitting between bound electronic surface states on liquid helium to resonance with the incident radiation. The $1 \rightarrow 2$ and $1 \rightarrow 3$ transitions are analogous to the Lyman-α and -β transitions of hydrogen.

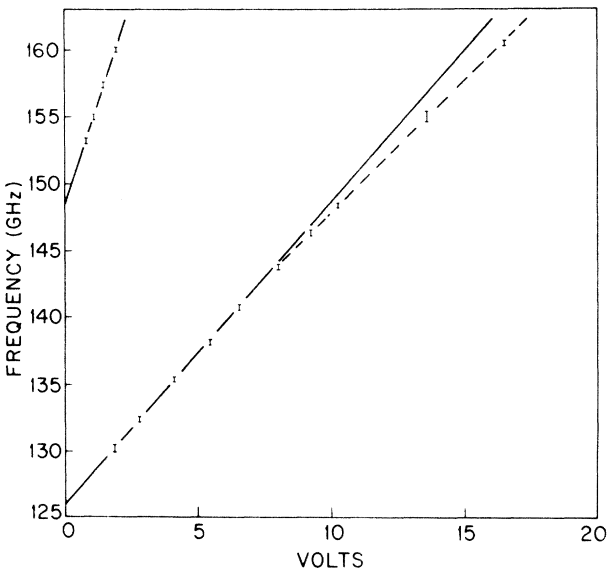

FIG. 2. Locus of measured resonance frequencies versus voltage across the experimental cell. Solid lines, least-squares fits to the lowest four data points. Their intercepts yield the frequencies of the unperturbed transitions and their slopes yield the tuning rates of the linear Stark effect. The intercepts are 125.9 ± 0.2 and 148.6 ± 0.3 GHz while the slopes are 2.3 and 5.9 GHz/V for the $1 \rightarrow 2$ and $1 \rightarrow 3$ transitions, respectively. The dashed line is merely to aid the eye.

olating to the zero-density limit, the unshifted line position is readily established to within ± 0.2 V. A plot of resonance frequency versus potential difference across the cell at resonance is presented in Fig. 2. Extrapolating the data points to $V = 0$ yields 125.9 ± 0.2 GHz for the unperturbed splitting between the ground and first excited states. Similarly, we obtain 148.6 ± 0.3 GHz for the splitting between the ground and second excited states. Both splittings are significantly larger than predictions based on the hydrogenic approximation. We shall discuss the causes of this after we finish reviewing the experimental data.

The slopes of the lines in Fig. 2 are 2.3 ± 0.1 GHz/V for the $1 \rightarrow 2$ transition and 5.9 ± 0.4 GHz/V for the $1 \rightarrow 3$ transition. The ratio of these slopes is 2.6 ± 0.2, in good agreement with 2.67 expected from the first-order perturbation calculation mentioned earlier.[12] The relative absorption cross sections of the resonances are in the ratio 1:5, in agreement with that expected for electric dipole transitions between the "hydrogenic" eigenstates.

The linewidth of the resonances has been studied over the very limited temperature interval from 1.2 to 1.5 K corresponding to vapor pressures from 0.5 to 3.6 Torr. The linewidth is observed to be 1.3 GHz at 1.0 Torr and appears to increase linearly with increasing vapor pressure. This is in agreement with estimates of the line broadening due to scattering from helium atoms in the vapor. More refined calculations are in progress.[13]

We now return to the differences between our extrapolated values of the eigenstate splittings and those predicted by the "hydrogenic" approximation. Presumably these are due to the oversimplified model of the potential used and therefore we modify it to obtain agreement with experiment. Although the apparently simplest modification to make to explain the difference is to lower the infinite barrier at the surface to some finite level V_0, in fact, the calculation of the eigenvalues in this case is obscured by the singularity in the $1/x$ potential (itself rather unphysical). Thus, for both computational simplicity and physical reasonableness, we shall consider two models from which these unphysical characteristics have been removed.

One is to take the potential to be $-Ze^2/x$ for $x \geq b$ but hold it constant at $-Ze^2/b$ for $0 < x \leq b$. This model has been considered by Cole.[2] The other model is merely to shift the origin of the image potential an amount b so that the potential

is $-Ze^2/(x+b)$ for $x>0$. Of these models the latter seems to us to be the most physically reasonable. We come to this conclusion by considering the microscopic causes of both the image potential and the potential step which occurs on entering the helium. The step is due to the overlap of the wave function of the extra electron with those of the electrons already on the helium atoms. The image potential is due to the polarization of the individual helium atoms by the extra electron. If we ignore the problems associated with both the liquid and quantum nature of the surface (reasonable since the electron averages over 10^4–10^5 atoms), then we would expect the origin plane of the image potential (located at the centers of the atoms in the first layer) to be deeper into the helium than the plane where the potential step occurs (located at the outer edge of the atoms) by a distance of order 1 Å.

To obtain agreement with experiment, taking V_0 to be 1 eV,[9] we find the necessary shift in the origin plane of the image potential is only 1 Å. In Cole's model, for the same V_0, a value of b equal to 10 Å is necessary, which seems unrealistically large.[14] In view of the striking differences between these two simple models we hope that more extensive and realistic calculations will be able to utilize this type of spectroscopic data to obtain new information about details of the surface of liquid helium which have been hitherto inaccessible to measurement.

In summary, we have observed transitions between bound electron states on the surface of liquid helium and verified their image potential character. The states display the expected linear Stark effect. In addition, we find the observed splittings agree well with a model in which the image potential originates 1 Å inside the helium surface.

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[12]Because of the finite size of the experimental cell it is difficult to convert accurately a potential difference into an electric field and thus we cannot compare the actual slopes in volts to the calculated ones in volts/centimeter to better than 15%.

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[14]We should point out that in these models V_0 can be traded off against b, so that, for example, in Cole's model a V_0 of 1.6 eV would correspond to a value of b equal to 3.6 Å.